

#### IN THE U.S. PATENT AND TRADEMARK OFFICE

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Applicant: Satoshi FURUTA

For: METHOD FOR MANUFACTURE OF ESTERS BY TRANSESTERIFICATION

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Commissioner for Patents P.O. Box 1450 Alexandria, VA 22313-1450

#### APPELLANT'S BRIEF ON APPEAL

Sir:

This is an appeal from the final rejection dated July 30, 2009 finally rejecting Claims 1, 3, 10, 12, 14-16, 18 and 19.

#### REAL PARTY IN INTEREST

Japan Energy Corporation is the assignee of the present application and the real party in interest.

#### RELATED APPEALS AND INTERFERENCES

There are no related appeals and interferences to the present application.

#### STATUS OF CLAIMS

Claims 1, 3, 10, 12, 14-16, 18 and 19 are pending and are the claims on appeal. Claims 2, 4-9, 11, 13 and 17 have been canceled.

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#### STATUS OF AMENDMENTS

An Amendment After Final Rejection has not been filed.

#### SUMMARY OF CLAIMED SUBJECT MATTER

Appellant's invention, as defined by independent Claim 1, is directed to a method for the manufacture of an ester by transesterification which comprises the step of bringing a starting material ester in a liquid phase state and an alcohol in a vapor phase state (paragraph [0019] of the clean copy of the substitute specification) into contact with an amorphous solid acid catalyst consisting of (A) an amorphous zirconium oxide and (B) aluminum oxide (paragraphs [0005] and [0006] of the clean copy of the substitute specification) wherein the content of the aluminum oxide is, calculated as the element, 25 to 1 wt.% based on the zirconium element weight (paragraph [0012] of the clean copy of the specification).

Appellant's invention, as defined by independent Claim 12, is directed to a method for the manufacture of an ester by transesterification comprising the step of bringing a starting material ester in a liquid phase state and an alcohol in a vapor phase state (paragraph [0005] of the clean copy of the specification) into contact with an amorphous solid acid catalyst consisting of (A) an amorphous zirconium oxide and (B) phosphorus oxide, wherein the content of the phosphorus oxide is, calculated as the element, 6 to 1 wt.% based on the zirconium element weight (paragraph [0014] of the clean copy of the specification).

Appellant's invention, as defined by independent Claim 16, is directed to a method for the manufacture of an ester by transesterification comprising the step of bringing a starting material ester in a liquid phase state and an alcohol in a vapor phase state (paragraph [0005] of the clean copy of the specification) into contact with an amorphous solid acid catalyst consisting of (A) an amorphous zirconium oxide and (B) titanium oxide wherein the content of the amorphous zirconium oxide in the catalyst is 40 to 90 wt.% and the

content of the titanium oxide is 60 to 10 wt.% in the catalyst (paragraphs [0005] and [0010] of the clean copy of the specification).

#### GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

The first ground of rejection to be reviewed on appeal is whether Claims 1, 3, 10, 12, 14 and 15 are unpatentable under 35 USC 103(a) over Ginosar et al. The second ground of rejection to be reviewed on appeal is whether Claims 16, 18 and 19 are unpatentable under 35 USC 103(a) over Bayense et al.

#### ARGUMENT

The presently claimed invention is directed to a method for manufacturing an ester by transesterification. In the present invention, a starting material ester in a liquid-phase state and an alcohol in a vapor-phase state are brought into contact with a solid acid catalyst consisting of either an amorphous zirconium oxide and aluminum oxide, an amorphous zirconium oxide and phosphorus oxide or an amorphous zirconium oxide and titanium oxide.

The present invention was arrived at in order to overcome conventional problems associated with transesterification reactions by providing a method which conducts a transesterification reaction in a short period of time and under a pressure on the order of a normal pressure. The catalyst used in the claimed reaction and the product produced by the reaction can easily be separated from each other, thereby making the inventive method more efficient. Furthermore, utilizing a catalyst consisting of an amorphous zirconium oxide and at least one other oxide selected from the group consisting of aluminum oxide, phosphorus oxide and titanium oxide allows the presently claimed transesterification reaction to achieve very high conversion rates.

# REJECTION OF CLAIMS 1, 3, 10, 12, 14 AND 15 OVER GINOSAR ET AL

The Ginosar et al reference is directed to a process for producing alkyl esters used in biofuels and lubricants through the transesterification of glyceride- or esterification of free fatty acid-containing substances in a single critical phase medium. In this reference, the input glyceride- or free fatty acid-containing substance is dissolved with an alcohol or water into a critical fluid medium, the glyceride- or free fatty acid-containing substance reacted with the alcohol or water input over either a solid or liquid acidic or basic catalyst and the products separated from each other and from the critical fluid medium.

In the final rejection, the Examiner has once again stated that Ginosar et al suggests the use of "alcohol steam" and that since the meaning of steam is vapor, Ginosar et al reads on the present claims. Appellant has thoroughly reviewed the Ginosar et al reference and cannot find any reference to "alcohol steam" and only finds reference to an input alcohol stream 102 in the disclosure of this reference.

In Ginosar et al, the reactants enter a reactor, whether batch or continuous, are dissolved in a critical fluid and the critical fluid forms a single-phase medium in which diffusion of the reactants into different liquid phases is eliminated and mass transfer limitations are essentially eliminated to thereby increase the overall reaction rate (column 2, lines 21-26 and column 5, lines 22-26). As stated in Claim 12 of this reference, the reaction occurs in a single phase to produce a final product.

In contrast to the Ginosar et al reference, the presently claimed invention requires that the reactants be in two different phases, an ester in a liquid phase and an alcohol in a vapor phase. As such, the reaction does not require a high pressure and be conducted under critical conditions as required in Ginosar et al. Since the reaction system of the present invention is completely different from that of Ginosar

et al, the results achieved by the present invention would not be obvious in view of this reference. Ginosar teaches the reaction occurring in a single phase while the present invention requires a reaction occurring between an ester in a liquid phase and an alcohol in a vapor phase.

As a catalyst used in the transesterification reaction of Ginosar et al, microporous crystalline solids, and noncrystalline inorganic oxides such as alumina, silica, silicaalumina, boria, oxides of phosphorus, titanium oxide, zirconium dioxide, cromia, zinc oxide, magnesia, calcium oxide, iron oxides, unmodified or modified with chlorine, fluorine, sulfur or an acid or base, as well as mixtures thereof or an exchange resin with either acidic or basic properties are disclosed as being used. However, there is no specific example in this reference using an amorphous catalyst including amorphous zirconium oxide or the specific effect of amorphous zirconium oxide over crystalline zirconium oxide as demonstrated in Tables 1-3 in the present specification. the final rejection, the Examiner states that "mere statements by the inventors are not supported by evidence". However, the Examples and Comparative Examples contained in the present specification constitute evidence as they are provided under the oath of the inventor. As such, they cannot be merely dismissed as being "arguments" of the inventor.

As shown in Table 1 in the present specification, the catalyst of test Examples 2-6 showed a high conversion rate than the catalyst of Examples 1 and 7 due to amorphous zirconium oxide being used. Likewise in Table 2 with test Examples 8-10 as compared to test Example 7 and Examples 11-23 in Table 3 as compared to test Example 7. Therefore, Appellant respectfully submits that not only does the Ginosar et al reference make a showing of prima facie obviousness under 35 USC 103(a) of Claims 1, 3, 10, 12, 14 and 15, the evidence of unobviousness of record in the present application is more than sufficient to overcome any proper 35 USC 103(a) rejection.

# REJECTION OF CLAIMS 16, 18 AND 19 UNDER 35 USC 103(a) AS BEING UNPATENTABLE OVER BAYENSE ET AL

The Bayense et al reference discloses a process for the transesterification of carboxylic acid esters in which a catalyst is used which is substantially insoluble in a reaction mixture under reaction conditions. The catalyst comprises at least one silicate of Group IVB elements. outset, the consisting essentially of language used in Claim 16 expressly excludes the critical silicate of this reference therefrom. Moreover, although this reference discloses the use of titanium-containing silica catalysts, there is no example in this reference of a catalyst using zirconium. Bayense et al additionally states that the silicates can either be crystalline silicates or amorphous silicates and does not show any preference between the two. As such, there is no teaching in this reference regarding the advantages of using amorphous zirconium oxide over crystalline zirconium oxide.

The superiority of the presently claimed amorphous zirconium-titanium catalyst of currently presented Claim 16 over a zirconium-titanium catalyst containing a crystallized part is shown in test Examples 2 and 19 in the present specification as compared to the partially crystallized catalyst of test Examples 1, 3-5, 18 and 20-22. The amorphous catalyst of the present invention having the specified components in the specified compositional range exhibited high conversion rates and enabled the production of the target esters with a good efficiency as compared to the catalysts which contain partially crystallized components. This is clearly unexpected in light of the prior art cited by the Examiner and further establishes the patentability thereover.

#### CONCLUSION

For the reasons advanced above, it is respectfully submitted that the presently claimed invention is patentably distinguishable over the prior art cited by the Examiner. Reversal of the Examiner is respectfully solicited.

Respectfully submitted,

Terryence F. Chapman

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Encl: Claims Appendix

Evidence Appendix

Related Proceedings Appendix

Postal Card

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#### CLAIMS APPENDIX

- 1. A method for the manufacture of an ester by transesterification comprising the step of bringing a starting material ester in a liquid phase state and an alcohol in a vapor phase state into contact with an amorphous solid acid catalyst consisting of (A) an amorphous zirconium oxide and (B) aluminum oxide, wherein the content of the aluminum oxide is, calculated as the element, 25 to 1 wt.% based on the zirconium element weight.
- 3. The method according to claim 1, wherein the starting material ester is an oil or fat and the alcohol is methanol or ethanol.
- 10. The method according to Claim 1, wherein the starting material ester is a glyceride ester of a saturated or unsaturated aliphatic carboxylic acid having from 8-24 carbon atoms.
- 12. A method for the manufacture of an ester by transesterification comprising the step of bringing a starting material ester in a liquid phase state and an alcohol in a vapor phase state into contact with an amorphous solid acid catalyst consisting of (A) an amorphous zirconium oxide and (B) phosphorus oxide, wherein the content of the phosphorus oxide is, calculated as the element, 6 to 1 wt.% based on the zirconium element weight.
- 14. The method according to Claim 12, wherein the starting material ester is an oil or fat, and the alcohol is methanol or ethanol.
- 15. The method according to Claim 12, wherein the starting material ester is a glyceride ester of a saturated or

unsaturated aliphatic carboxylic acid having from 8-24 carbon atoms.

- 16. A method for the manufacture of an ester by transesterification comprising the step of bringing a starting material ester in a liquid phase state and an alcohol in a vapor phase state into contact with an amorphous solid acid catalyst consisting of (A) an amorphous zirconium oxide and (B) titanium oxide, wherein the content of the amorphous zirconium oxide in the catalyst is 40 to 90 wt.% and the content of the titanium oxide is 60 to 10 wt.% in the catalyst.
- 18. The method according to Claim 16, wherein the starting material ester is an oil or fat, and the alcohol is methanol or ethanol.
- 19. The method according to Claim 16, wherein the starting material ester is a glyceride ester of a saturated or unsaturated aliphatic carboxylic acid having from 8-24 carbon atoms.

## EVIDENCE APPENDIX

There is no extrinsic evidence being relied upon by the Appellant.

## RELATED PROCEEDINGS APPENDIX

There are no related proceedings to the present application.